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The role of interface in improving fracture toughness of shaped steel fiber-reinforced composites

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Abstract

Mechanical properties of composite materials and their final performance are strongly influenced by the structure and properties of the fiber–matrix interface. A balance between sufficient load transfer and fiber pullout properties helps improve the performance of the composite materials. In this research, we employed sulfuric acid treatment to enhance fiber pullout and debonding through decreasing the fiber–matrix interlock. Atomic force microscope and scanning electron microscopy were employed to investigate the surface morphology of both treated and untreated steel fibers. Our results indicate that surface treatment improves the fiber roughness and therefore hinders brittle failures of the epoxy fiber-reinforced composite materials.

Keywords

Short fiber, epoxy composite, surface morphology, crack deflection, interface

Introduction

Composite materials can provide a unique combination of electrical, mechanical, thermal, chemical, or biological properties[1] with wide applications in the aerospace, automotive, and biomedical industries.[2–6] Various categories of the matrices and the reinforcements are employed to construct composite materials with desired properties and functionality. Among these, epoxy polymers are one of the most commonly used matrices in many different categories of composite materials. Epoxy polymers have high chemical and heat resistance, good electrical properties, easy processability, good-to-excellent mechanical properties, and low shrinkage upon cure.[7–10]

Reinforcements are either continuous or discontinuous and, depending on their aspect ratio, they can be categorized into particles, fibers, or sheets. Fiber-reinforced composites are categorized into two main groups, referred to as short fiber-reinforced composites and long fiber-reinforced composites. In comparison with the long fiber-reinforced composites, short fiber-reinforced composites can be fabricated into complex shapes using low-cost manufacturing methods, such as injection molding, compression molding, or extrusion at high production rates. Furthermore, variety in selection of the matrices with different thermal, mechanical, and chemical properties is another advantage of short fiber composite materials over continuous fiber composite materials.[11–14] In this research, we are focused on the ductile short fiber-reinforced epoxy-based composite materials.

It is well established that in addition to the importance of size, loading, aspect ratio, dispersion state, and alignment of the reinforcements as well as physical and chemical properties of the matrices, the properties of polymer composites are influenced by the interface region between the reinforcements and the matrices.[13,15–17] Bonding between the fiber and the matrix

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is influenced by different mechanisms. Adsorption, wetting, electrostatic attraction, chemical bonding, reaction bonding, and mechanical bonding are dominant mechanisms affecting the interface's adhesion. The interface condition can be altered through modifying mechanical or chemical properties of the fiber's surface.[18–22] Surface treatment and geometrical shaping were also used to exploit inherent fiber plasticity to improve toughness of composite materials.[22–28] Geometrical shaping can be successfully employed to anchor the fiber into the matrix, which leads to a higher contribution of the fibers to the composite toughness. If fibers easily pull out of the matrix, their plastic work is not utilized to improve fracture toughness of the composite. Therefore, one might conclude that geometrical shape of the fiber is as important as the properties of the matrix.[29–31] Surface treatment, on the other hand, has been employed to improve interfacial adhesion or to make a surface-roughened fiber.[21,32,33] For example, addition of functional groups or using coupling agents such as silane can improve the interfacial adhesion between fiber and matrix while sulfuric acid treatment can produce a rougher surface.[21,32–34] In addition, recent research has shown that surface treatment is an effective technique for changing the microstructure at the fiber–matrix interface in both laminate and fiber-reinforced composites, and can improve their final performance.[35]

Behavior of a crack in the interface, affected by the properties of the interface, plays an important role in the final performance of the composite material. When increasing tensile load in the direction of the fiber, a microcrack grows until it reaches the fiber–matrix interface. At this stage, there are at least three possible paths for a crack to grow. In case of poor interfacial bonding, a crack is deflected along the interface. Therefore, the fiber remains intact and only frictional force during the fiber pullout as well as debonding processes play a role in improving the fracture toughness of the composite. These mechanisms adversely affect the composites' strength. However, if the interface is too strong, a crack penetrates the fiber, as it reaches the fiber, and does not propagate along the interface. As a result, before fiber debonding takes place, the fiber breaks and has no chance to develop plastic deformation and dissipate energy. On the contrary, a strong interfacial bonding can lead to an efficient load transfer and consequent increases in the strength of composite materials. A strong interface usually acts against crack propagation and improves rigidity of the composite material. Nevertheless, optimum properties of the fiber-reinforced composites are believed to be achieved by a weak-to-moderate interfacial bonding or a deformable interphase and an efficient load transfer mechanism.[36]

Experimental and material

Poly(bisphenol A-*co*-epichlorohydrin), glycidyl end-capped epoxy, *n*-butyl glycidyl ether, and cycloaliphatic polyamine were obtained from Aldrich, Germany. *N*-butyl glycidyl ether and polyamine have linear formulas of $C_7H_{14}O_2$ and $H_2NC_6H_7(CH_3)_3CH_2NH_2$, respectively. Carbon steel fibers were purchased from China National Building Materials Corporation, China. Sulfuric acid and acetone were purchased from Merck, Germany. To prepare the polymer mixture, 90 wt% of poly(bisphenol A-*co*-epichlorohydrin) was mixed with 10 wt% of cycloaliphatic polyamine to reduce the viscosity. Then, approximately 23 wt% of the catalyst was added to the epoxy polymer mixture for the proper curing of the matrix. Material properties are shown in Table 1. According to XRD analysis, carbon steel fibers contain 0.05 wt% C, 0.18 wt% Si, 0.44 wt% Mn, 0.031 wt% P, 0.028 wt% S, 0.04 wt% Cr, 0.003 wt% Al, 0.02 wt% Cu, and 0.02 wt% Ni.

In this research, the pullout force was measured using single fiber pullout test with the fiber's embedded length of 7.5 mm for both treated and untreated fibers. To perform tensile tests all composite samples were reinforced with 16 wt % of the zigzag shaped treated or untreated fibers. Tensile specimens shape is shown in Figure 1, which is according to ASTM D638-02, Type I. A minimum of five tests was performed for both tensile and pullout tests. To remove the air bubbles from

Table 1. Materials' properties.

Materials Properties	Fiber	Matrix
Young's modulus (GPa)	135.80	2.05
Poisson's ratio	0.28	0.30
Diameter (mm)	0.5	–
Volume fraction	0.03%	0.97%
Weight fraction	16%	84%

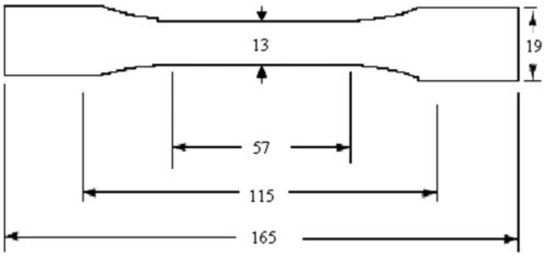

Figure 1. Schematic of the composite samples used for the tensile tests; shape and dimensions are according to ASTM D638-02, Specimen Type I.

the epoxy matrix, resin mixture was centrifuged after the resin was mixed with the curing agent. All samples were cured at room temperature for the total time of 48 h. Selection of room temperature in this research should not be interpreted as the ideal temperature to improve the adhesion between fiber and matrix. To find the best adhesion between fiber and matrix one might consider to study the effect of temperature on interface properties.[37] The strength of composite samples was measured using a Zwick 1446-60 tensile tester with 10 kN load cell at a crosshead speed of 5 mm/min.

To prepare the composite samples for tensile test, initially a negative mold was cut using Teflon material. Then, a silicon rubber mold was made from the Teflon mold. After mixing the proper ratio of the resin and the curing agent, half of the required mixture was poured in the silicon mold and cured for 24 h. In the next step, reinforcing fibers as well as the remainder of the resin mixture were added to the mold. Another 24 h of curing was required before samples could be removed from the mold. After composite samples were removed from the mold, dimensions of each sample were measured using a micrometer.

For the surface modification, a sulfuric acid etching technique was used. Treatment was carried out for sulfuric acid with different concentrations (i.e. 5, 10, 20, and 40 wt%) and according to atomic force microscope (AFM) results 10 wt% was the optimal concentration of sulfuric acid. RMS surface roughness of the fibers after treatment with 5, 10, 20, and 40 wt% sulfuric acid are 145, 343, 67.3, and 99.8 nm, respectively. All the fibers were treated by immersing in sulfuric acid solution for 5 min and were immediately rinsed with NaOH solution, to prevent rusting, and finally were dried using compressed air.

Results and discussion

Although transverse properties of the composite materials significantly depend on the tensile strength of the interface, having a strong interface is not always favorable. Strong interfaces result in penetration of the cracks into the fiber and consequent brittle fracture. On the other hand, a composite material with a moderate-strength interface can provide sufficient load transfer to the fibers while prohibiting brittle fracture of the composite. Toughness of composite materials is affected by different failure mechanisms such as matrix fracture, fiber fracture, fiber–matrix interface debonding, postdebonding friction, stress redistribution, fiber pullout, crack deflection, and microcracking. Contribution of each of the previously mentioned mechanisms on the final composite fracture toughness is defined by the relative strength of the interface to the strength of the reinforcing fibers and matrix. A moderate-strength interface is required for most of the previously mentioned mechanisms to take place.[21,29,32,38]

Results of the tensile test on our composite samples reinforced with untreated fibers reveal very brittle fractures, with crack penetration into the fibers being the dominant mechanism in the interface. To avoid penetration of the cracks into the fibers, which enhances the composite fracture toughness, we employed an etching technique to modify the fiber's roughness and consequently alter the behavior of the crack in the fiber–matrix interface. Applications of sulfuric acid treatment, as an etching technique, on the rough surface of the fibers resulted in a decrease of surface roughness. Such a decrease in the roughness is due to higher rate of corrosion at the area with higher surface energy on the rough surface of the fibers.

As shown in the scanning electron microscopy (SEM) and AFM graphs of the fibers (see Figures 2 and 3), the surface of untreated fibers has rougher features than those of acid-treated fibers. Therefore, micromechanical interlocking between fibers and matrix is not as strong in the treated fiber-reinforced composites compared to the untreated ones and results in reduction in fiber–matrix interface strength.

To investigate the effects of surface modification on the strength of the interface, pullout tests as well as

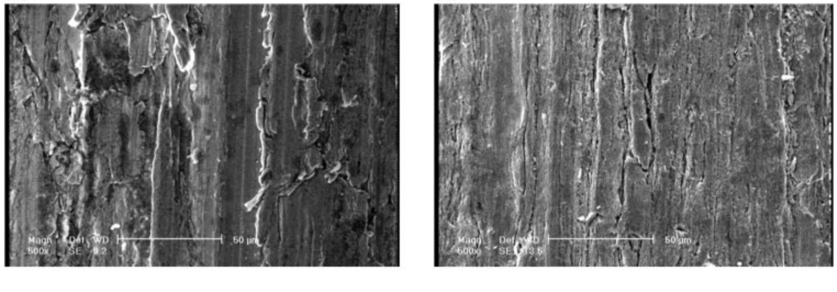

Figure 2. SEM picture of (a) untreated fibers and (b) acid-treated fibers.
SEM: scanning electron microscopy.

tensile tests were carried out for both treated and untreated fibers (shown in Figure 4). From the pullout test, it is clear that treated fibers require less force in order to be pulled out of the matrix. That is a direct consequence of weaker interlocking between the fiber and matrix due to the changes in surface microstructure after acid treatment. Figure 4(b) and Table 2 show performance and mechanical properties of the composite reinforced with untreated and treated fiber, respectively. Sulfuric acid treatment results in a significant increase in fracture toughness of the composite (124%) samples; however, the strength of the samples decreases 11%.

Reported fracture toughnesses are the sum of the fracture toughness before and after the point of maximum applied load. Here we refer to the possible mechanisms contributing to the overall improvement in the total toughness when samples are reinforced with treated fibers. Comparison between treated and untreated fiber-reinforced composites reveals that resistance against the growth of a crack through the interface decreases upon treatment of the fibers. Therefore, when treated fibers are used for reinforcing composites, distributions of cracks in the matrix, as well as along the interface, are more dominant than crack penetration in the fibers. As a crack deflects along the interface, interfacial debonding and frictional sliding are more likely to happen, which results in enhancing the fracture toughness of the composite samples. Crack deflection along the interface creates more fracture surface area which can improve composites' fracture toughness, as a result of specific energies absorbed during creation of the new surfaces (see Figure 5(a) and (b)). The energy dissipated when steel fibers lose their zigzag shape during the pullout process is another mechanism that possibly contributes in improving fracture toughness of the composite samples (see Figure 5(c)). Moderate strength of the interface in the treated fiber-reinforced composite provides sufficient load transfer to the fibers and also results in the fracture of some of the fibers (see

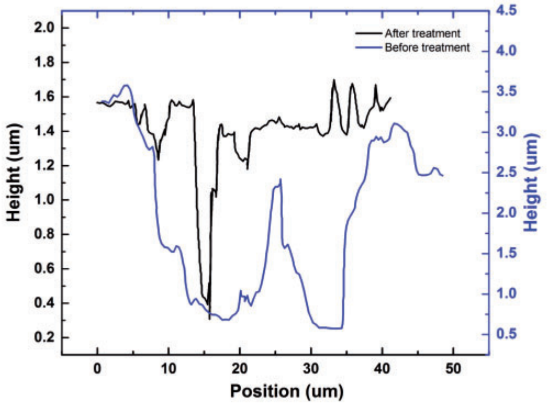

Figure 3. AFM profile of untreated and sulfuric acid treated fibers; concentration of sulfuric acid is 10 wt%.
AFM: atomic force microscope.

Table 2. Young's modulus, fracture toughness, and strength of composites reinforced with untreated and treated waved fibers.

	Properties		
Specifications	Young's modulus (GPa)	Fracture toughness (kJ/m)	Strength (MPa)
Treated fiber	3.4	43.59	19.21
Untreated fiber	3.4	19.41	21.61

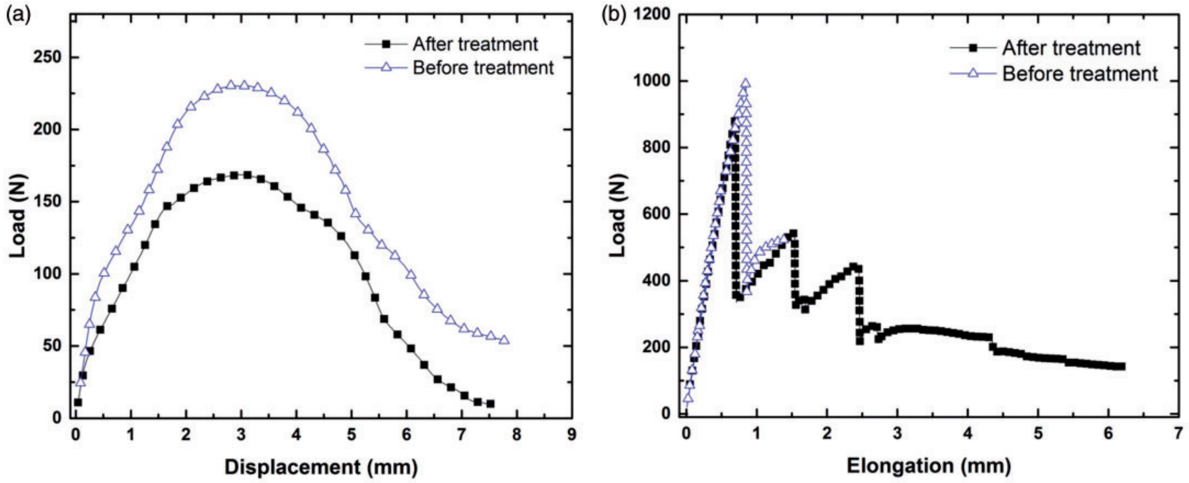

Figure 4. (a) Pullout test for treated and untreated fiber-reinforced composite samples, embedment depth of fibers is 7.5 mm. (b) Load–elongation behavior of treated and untreated waved fiber-reinforced composite under tensile test.

Figure 5). Therefore, fibers' ductility can contribute to improve the final ductility of these composite samples. However, to quantify the contribution of fibers' ductility as well as change in the fiber's shape on the overall composite toughness more experiments are required.

In the untreated fiber composites, fibers fracture due to crack penetration is the most dominant mechanism contributing to composite toughening (see Figure 5(d)) but fiber fracture does not provide high damage tolerance. Force–elongation behavior of treated and untreated fiber-reinforced composite (Figure 4(b)) confirms penetration of the cracks into fibers when the fibers are not treated.

SEM image (shown in Figure 6) of the fiber–matrix interface reveals that after fiber treatment, cohesive and interfacial bond failure are more dominant in the interface.[33] According to the SEM of the treated fiber, after the composite is fractured (Figure 6(a)), crack deflection in the interface left some fragments of the matrix on the fiber's surface. These fragments, together with the zigzag shape of the fibers, act as anchors which restrict the debonded fibers from pulling out of the

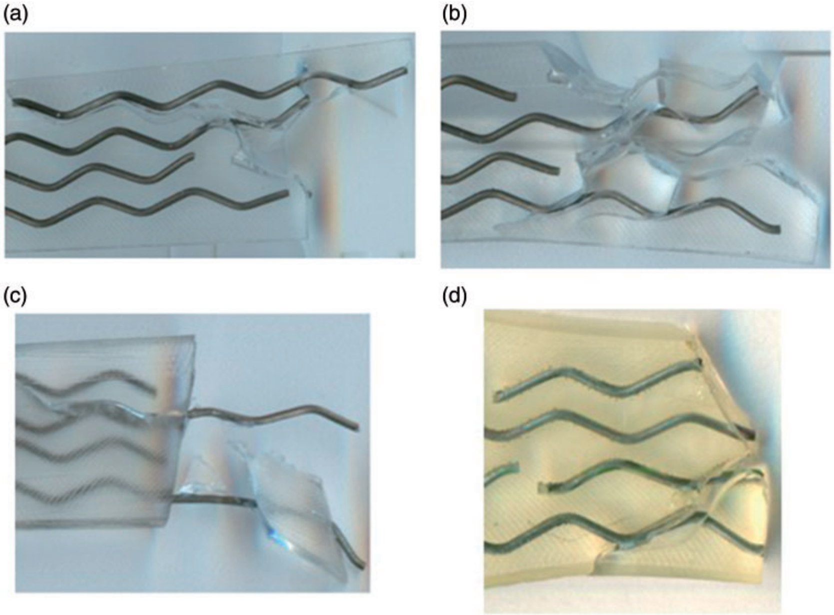

Figure 5. Crack behavior in treated (a), (b), and (c) and untreated (d) fiber-reinforced composite samples. (a) and (b) clearly show the propagation of the crack in the interface while (c) presents how fibers lose their zigzag shape during the fracture. (d) shows the penetration of the crack in the fibers which lead to fibers breakage rather than pullout.

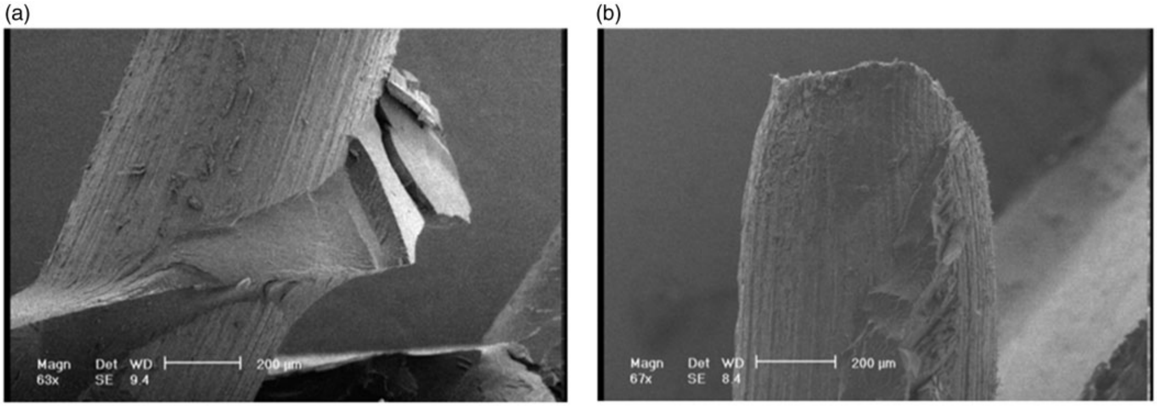

Figure 6. SEM micrograph of interface in the treated fiber composites after fracture. (a) When a crack deflects and (b) when a crack deflects at interface initially and finally penetrates the fiber.
SEM: scanning electron microscopy.

matrix and results in increases in postdebonding friction work.

Conclusion

In this work, sulfuric acid treatment is employed as an etching technique to reduce the surface roughness of the steel fibers. Due to high surface roughness, untreated fibers have strong interlocking with the matrix which resulted in brittle fracture of the composite samples. Epoxy polymer reinforced with modified fibers provided us with a moderate strength interface. Producing moderate strength interfaces between the fiber and matrix results in toughness improvement while we have sufficient load transfer to the fibers. Toughness improved as a result of crack propagation at the interface, fiber pullout, postdebonding friction work, as well as fiber elongation. However, to make a broad conclusion about the effect of the surface treatment on mechanical properties of the composite, such treatment should be examined with different zigzag angles as well as curing temperatures.

Declaration of Conflicting Interests

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References

1. Al-Oqla FM, Sapuan SM, Anwer T, et al. Natural fiber reinforced conductive polymer composites as functional materials: a review. *Synth Met* 2015; 206: 42–54.
2. Agrawal A, Rahbar N and Calvert PD. Strong fiber-reinforced hydrogel. *Acta Biomater* 2013; 9: 5313–5318.
3. Rezwan K, Chen QZ, Blaker JJ, et al. Biodegradable and bioactive porous polymer/inorganic composite scaffolds for bone tissue engineering. *Biomaterials* 2006; 27: 3413–3431.
4. Stankovich S, Dikin DA, Dommett GHB, et al. Graphene-based composite materials. *Nature* 2006; 442: 282–286.
5. Mohammad M and Winey KI. Polymer nanocomposites containing carbon nanotubes. *Macromolecules* 2006; 39: 5194–5205.
6. Goldberg AJ and Burstone CJ. The use of continuous fiber reinforcement in dentistry. *Dent Mater* 1992; 8: 197–202.
7. Allaoui A, Bai S, Cheng HM, et al. Mechanical and electrical properties of a MWNT/epoxy composite. *Compos Sci Technol* 2002; 62: 1993–1998.
8. Zhu J, Peng H, Rodriguez-Macias F, et al. Reinforcing epoxy polymer composites through covalent integration of functionalized nanotubes. *Adv Funct Mater* 2004; 14: 643–648.
9. Yu A, Ramesh P, Itkis ME, Bekyarova E, et al. Graphite nanoplatelet-epoxy composite thermal interface materials. *J Phys Chem C* 2007; 111: 7565–7569.
10. Bekyarova E, Thostenson ET, Yu A, et al. Multiscale carbon nanotube-carbon fiber reinforcement for advanced epoxy composites. *Langmuir ACS J Surf Colloids* 2007; 23: 3970–3974.
11. Adekunle K, Dan A and Skrifvars M. Biobased composites prepared by compression molding with a novel thermoset resin from soybean oil and a natural-fiber reinforcement. *J Appl Polym Sci* 2010; 116: 1759–1765.
12. Bledzki AK and Gassan J. Composites reinforced with cellulose based fibers. *Prog Polym Sci* 1999; 24: 221–274.
13. Herrera-Franco PJ and Valadez-González A. A study of the mechanical properties of short natural-fiber reinforced composites. *Compos Part B Eng* 2005; 36: 597–608.
14. Fonseca RB, De Almeida LN de, Mendes GAM, et al. Effect of short glass fiber/filler particle proportion on flexural and diametral tensile strength of a novel fiber-reinforced composite. *J Prosthodont Res* 2016; 60: 47–53.
15. Moon CK. Study on the interfacial properties of three-dimensionally arranged glass fiber/epoxy resin model composites. *J Appl Polym Sci* 2010; 2: 1483–1490.
16. George J, Sreekala M and Thomas S. A review on interface modification and characterization of natural fiber reinforced plastic composites. *Polym Eng Sci* 2001; 41: 1471–1485.
17. Pan Y, Weng GJ, Meguid SA, et al. Interface effects on the viscoelastic characteristics of carbon nanotube polymer matrix composites. *Mech Mater* 2013; 58: 1–11.
18. Almeida RS, Damato CA, Botelho EC, et al. Effect of surface treatment on fatigue behavior of metal/carbon fiber laminates. *J Mater Sci* 2008; 43: 3173–3179.
19. Varga C, Miskolczi N, Bartha L, et al. Improving the mechanical properties of glass-fibre-reinforced polyester composites by modification of fibre surface. *Mater Des* 2010; 31: 185–193.
20. Chen H, Jia C and Li S. Interfacial characterization and thermal conductivity of diamond/Cu composites prepared by two HPHT techniques. *J Mater Sci* 2011; 47: 3367–3375.
21. Ochoa-Putman C and Vaidya UK. Mechanisms of interfacial adhesion in metal–polymer composites – effect of chemical treatment. *Compos Part A Appl Sci Manuf* 2011; 42: 906–915.
22. Zaldivar RJ, Kim HI, Steckel GL, et al. Surface preparation for adhesive bonding of polycyanurate-based fiber-reinforced composites using atmospheric plasma treatment. *J Appl Polym Sci* 2011; 120: 921–931.
23. Chen W, Tao Z, Fan L, et al. Effect of poly (etherimide) chemical structures on the properties of epoxy/poly (etherimide) blends and their carbon fiber-reinforced composites. *J Appl Polym Sci* 2011; 119: 3162–3169.
24. Jancar J. Review of the role of the interphase in the control of composite performance on micro- and nano-length scales. *J Mater Sci* 2008; 43: 6747–6757.
25. Garcia FG, Soares BG, Leyva ME, et al. Influence of aliphatic amine epoxy hardener on the adhesive properties of blends of mono-carboxyl- terminated poly

(2-ethylhexyl acrylate-co-methyl methacrylate) with epoxy resin. *J Appl Polym Sci* 2010; 117: 2762–2770.

26. Wetherhold RC and Bös J. Ductile reinforcements for enhancing fracture resistance in composite materials. *Theor Appl Fract Mech* 2000; 33: 83–91.
27. Bagwell RM and Wetherhold RC. Debond behavior of copper fibers in thermoset matrices and their effect on fracture toughening. *J Adhes Sci Technol* 2003; 17: 2223–2242.
28. Ma K, Chen P, Wang B, et al. A study of the effect of oxygen plasma treatment on the interfacial properties of carbon fiber/epoxy composites. *J Appl Polym Sci* 2010; 118: 1606–1614.
29. Bagwell RM and Wetherhold RC. Improvement in fracture toughness of an epoxy/copper composite through the use of various end shaped fibers. *Mater Sci Eng A* 2003; 361: 294–301.
30. Bagwell RM and Wetherhold RC. Fiber pullout behavior and impact toughness of short shaped copper fibers in thermoset matrices. *Compos Part A Appl Sci Manuf* 2005; 36: 683–690.
31. Bagwell RM and Wetherhold RC. End-shaped copper fibers in an epoxy matrix – predicted versus actual fracture toughening. *Theor Appl Fract Mech* 2005; 43: 181–188.
32. Callens MG, Gorbatikh L, Bertels E, et al. Tensile behaviour of stainless steel fibre/epoxy composites with modified adhesion. *Compos Part A Appl Sci Manuf* 2015; 69: 208–218.
33. Wetherhold RC, Corjon M and Das PK. Multiscale considerations for interface engineering to improve fracture toughness of ductile fiber/thermoset matrix composites. *Compos Sci Technol* 2007; 67: 2428–2437.
34. Wetherhold RC and Harry Z. A rapid chemical method for improving peel strength at Cu-epoxy interfaces. *Theor Appl Fract Mech* 2010; 53: 42–46.
35. Reddy MG, Nowshoba P and Gowd GH. Synthesis and characterization of *Hardwickia binata* fiber with epoxy. *Appl Mech Mater* 2014; 592–594: 874–878.
36. Kim JK and Mai YW. *Engineered interfaces in fiber reinforced composites*. Oxford: Elsevier, 1998.
37. Garcia FG, Leyva ME, Oliveira MG, et al. Influence of chemical structure of hardener on mechanical and adhesive properties of epoxy polymers. *J Appl Polym Sci* 2010; 117: 2213–2219.
38. Wetherhold RC and Bös J. Ductile reinforcements for enhancing fracture resistance in composite materials. *Theor Appl Fract Mech* 2000; 33: 83–91.